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TITLE FISSION PROPERTIES AND PRODUCTION MECHANISMS FOR THE HEAVIEST KNOWN ELEMENTS

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The Radiochemistry Group at Los Alamos has been particularly interested in the production and properties of the heavy elements at least as long ago as 1952 when the new elements, einsteinium (99) and fermium (100) were rather unexpectedly produced and identified in the debris from the Mike thermonuclear test. These elements were identified via the isotopes 20.5d ^{253}Es and 20.1-hour ^{255}Fm, representing capture of 15 and 17 neutrons respectively in ^{238}U. The longest-lived isotope of plutonium, 82-million year ^{244}Pu was also discovered on that test. We continued to look for other new heavy elements on subsequent tests in the Pacific and although many neutron rich heavy isotopes were produced and studied, including ^{245}Pu-^{245}Am, ^{246}Pu-^{246}Am, no elements above Z>100 were detected.

In the 1960's, both Livermore and Los Alamos fielded several underground tests at the NTS designed to enhance heavy element production, but again, no new elements were produced and the heaviest isotope identified was 100-day ^{257}Fm, regardless of the target material. In fact, ^{238}U still seemed to be the target of choice. At that time most of us were extremely puzzled by our failure to find ^{258}Fm or ^{259}Fm as we were not yet aware of the SF "disaster" which occurs at ^{258}Fm. We expected the fission half lives to be long enough to detect in nuclear test debris, or that these nuclides might even , decay to mendelevium (101) which had been discovered in 1955 as 76-min ^{256}Md. However, although relatively large amounts of heavy element isotopes were produced, we appeared to have reached a dead end at ^{257}Fm. Subsequent measurements by Hulet et al.[1] showed that the $T_{1/2}$ of ^{258}Fm was only 0.38 ms which explained why we couldn't find it although many of us still felt the odd mass nuclide, ^{259}Fm, should have a longer half life. However, an upper limit of 5 hours was placed on the SF half life for ^{259}Fm or ^{259}Md from our measurements on those tests.[2] So that was the bad news! The good news was that we were able to isolate enough ^{257}Fm (only ~1 SF/min) to make the first measurements of the mass yield curve for its spontaneous fission by the double kinetic energy method. (^{257}Fm decays principally by α emission but has a 0.2% SF branch.) These measurements were repeated using a source of ^{257}Fm produced in the HFIR reactor at Oak Ridge and we confirmed the fact that [illegible] greatly increased yield of [illegible] mass division over that previously observed for any SF or low energy fission process. These results were published[3] in 1971 and caused what might be called a "renaissance" of interest in SF and low energy fission because it was previously believed that all spontaneous and low energy fission resulted in highly asymmetric mass division. However, many of us felt that the enhanced yields for symmetric mass division in Fm might be expected and were due to the fact that Fm with $Z = 100$ could split symmetrically into two $Z = 50$ closed proton shell nuclei with neutron numbers approaching the closed $N = 82$ shell as the mass of the fissioning fermium isotope increases. Thus still higher yields of symmetric mass division should result for heavier fermium isotopes.

Finally, in 1975, Hulet and co-workers from Lawrence Livermore Laboratories and Wilhelmy, Weber and I at Los Alamos were able to produce[4] ^{259}Fm via the (t,p) reaction at the Los Alamos Van de Graaff on a target of 10^9 atoms of ^{257}Fm. The half life for ^{259}Fm was only 1.5 seconds (thus continuing the SF disaster) and its SF showed an extremely narrow, symmetric mass distribution with an unusually high total kinetic energy (TKE) of ≈240 MeV, a value which approaches the Q value for fission of ≈250 MeV. We have also measured[5] the SF of ^{258}Fm via electron-capture decay of 43-minute ^{258}Md produced by the ^{255}Es (α,n) reaction. The measured mass yields for Fm isotopes are shown in Figure 1. Recently, we have measured[6] the neutron deficient isotopes, ^{246}Fm and ^{248}Fm, and find that they exhibit highly asymmetric distributions (Figure 2.) We produced[5] the new 12.3-minute ^{256}Cf from the (t,p) reaction on ^{254}Cf and mass yields for the Cf isotopes are shown in Figure 3. Although ^{256}Cf has the same number of neutrons as ^{258}Fm, it does not show a similar abrupt change to symmetric fission. Neither is its TKE unusually high. A plot of TKE vs. $Z^2/A^{1/3}$ is shown in Figure 4. Only ^{258}Fm and ^{259}Fm appear to be "abnormal". Contour plots of TKE vs. mass fraction for ^{256}Fm, ^{257}Fm, and ^{259}Fm are shown in Figure 5.

Results of Hulet et al.[7] for ^{259}Md (also with 258 neutrons) indicate that although its mass distribution is broadly symmetric (Figure 6) its TKE is only ≈200 MeV, a "normal" value (Figure 7). The very high TKE for ^{258,259}Fm can be explained on the basis of coulomb repulsion which will be a maximum for the spherical fragments resulting from approach of the fragments to the doubly magic, spherical ^{132}Sn configuration. Since the energy released in binary fission is manifested as kinetic energy or excitation energy of the fragments which then can de-excite by neutron and gamma emission, very little energy is left for neutron emission from the near-spherical fragments formed by symmetric fission of ^{258}Fm and ^{259}Fm. Again, this is a marked deviation from the systematics. As shown in Figure 8, the average number of neutrons per fission, $\bar{\nu}$, for low energy fission generally increases with Z. We have made measurements[8] of $\bar{\nu}$ as a function of fragment TKE and mass ratio for the SF of ^{250}Cf, ^{252}Cf, ^{254}Cf, ^{256}Fm and ^{257}Fm. As expected, neutron emission decreases monotonically with increasing TKE for a given mass split. A comparison of the multiplicity distributions[9] for the highest TKE events from SF of ^{250}Cf, ^{252}Cf, and ^{257}Fm is shown in Figure 9 and illustrates the large probability for emission of 0 neutrons for ^{257}Fm for events with TKE > 240 MeV. This is consistent with a large fraction of these fragments being nearly spherical with resultant high TKE's which are nearly equal to the Q value. Similarly, we expect that in the case of SF of ^{258}Fm and ^{259}Fm where the TKE is about 240 MeV, close to the estimated Q values of around 250 MeV, neutron emission will be low. The fragments must be nearly spherical and only have sufficient excitation energy to emit about 1 neutron on the average. Perhaps ^{259}Md, which has one more proton but the same number of neutrons as ^{258}Fm, is a "transition" nucleus similar to ^{257}Fm and has more deformed fragments which will have sufficient excitation energy to emit several neutrons, and thus account for the 40 MeV difference in TKE. It has also been postulated that charged particle emission at scission might account for this deficit, but preliminary experiments[7] set a very low limit (1) on the abundance of such events. We hope to perform measurements of neutron emission for these two nuclides to see if that can account for the missing energy! Due to our recent evidence[10] for a transfer reaction in which ^{259}Fm was made with a cross section of about 15 nb by bombardment of ^{248}Cm with ^{18}O, an effective transfer of ^{11}Be, such "on-line" measurements now appear feasible. Although the cross section is not nearly as high as for the (t,p) reaction, much more target material is available and we were able to make much better measurements of the mass and kinetic energy distributions for ^{259}Fm with less interference from 2.6-h ^{256}Fm. Although the properties measured for the 1.5-second activity produced in this reaction (See Table I), indicate that it is most likely ^{259}Fm, an unknown isotope of Md cannot be ruled out. However, simple calculations[11] based on energy considerations favor ^{259}Fm production.

We have now made radiochemical measurements[12] of actinide yields for bombardment of ^{248}Cm with ^{16}O, ^{18}O, ^{20}Ne, and ^{22}Ne (Figures 10 and 11), and these results show that nearly all possible products between target and compound nucleus can be produced. Comparisons of the yields indicate that, in general, the maximum yield for a given element occurs about two mass units heavier for ^{18}O and ^{22}Ne than for ^{16}O and ^{20}Ne, respectively, reflecting the neutron excess of the projectile. The dependence of the yields on the energy of the projectiles is now being investigated. Preliminary analysis of the data shows different energy dependences for production of various actinide isotopes. These excitation functions appear to be consistent with the calculations based on simple energy balance considerations for the appropriate transfer reactions, assuming the energy of the projectile in excess of the coulomb barrier is apportioned to the target nuclei according to the fraction of the projectile mass transferred. Such transfer reactions give promise of providing a means of producing new neutron-rich isotopes for study and for tailoring the target-projectile system and energy to enhance the yield of the desired product. They may even provide a means of getting to the neutron-rich side of the elusive superheavy element region by bombardment of ^{248}Cm with ^{48}Ca ions. Figure 12 shows the energy available for the heavy product for reactions at the coulomb barrier and indicates additional projectile energy will be required for products with $Z < 114$.

Recently German scientists at GSI have reported[13] finding evidence for production of element 107 in the bombardment of ^{209}Bi with ^{54}Cr ions at an energy just above the coulomb barrier by the reaction $^{209}Bi + ^{54}Cr \rightarrow ^{262}107 + n$. They have detected 6 alpha decays (~ 10.4 MeV; $T_{1/2} \approx 5$ ms) which they attribute to $^{262}107$, based on α-α correlation measurements with the known daughters, $^{258}105$ and $^{256}103$. Unfortunately, with the velocity filter system, SHIP, which they use, they can only detect products in a very small angle about the beam direction, and thus they are restricted to compound nucleus reactions followed by isotropic particle evaporation. Therefore, transfer reaction products cannot be investigated since they will be distributed at larger angles to the beam direction. However, Münzenberg et al.[13] believe the reaction of ^{48}Ca with ^{248}Cm at energies near the coulomb barrier will produce the compound system $^{296}116$ at an excitation energy of only 2[illegible] MeV. Thus there may be some hope of forming a superheavy isotope by evaporation of only a few neutrons with cross sections of the order of a few tenths of a nb.

Investigations to date for transfer reactions and energy balance calculations indicate that bombardment of ^{248}Cm with ^{48}Ca at 15-20 MeV above the barrier might produce a desired superheavy product such as $^{288}112$ or $^{291}113$ (transfer of ^{40}S or ^{43}Cl) with an excitation of only a few MeV because of the negative Q values for these reactions. (See Figure 12) This allows use of higher energy particles which should increase the probability of large transfers while still keeping the product excitation energy low enough to minimize destruction by prompt fission, and might result in considerably higher production cross sections. Now the challenge is to devise systems for measuring the Z and A for such short-lived nuclides which may decay by SF rather than alpha emission, and to detect transfer products which do not move along the beam direction.

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Table I. Spontaneous fission properties of some heavy-element isotopes (taken from Ref. 10).

Nuclide	SF $T_{1/2}$ (s)	Peak-to valley ratio [a]	Pre-neutron TKE^b (MeV)	σ_{TKE}
^{250}Cf	5.4×10^{11}	>300 (RC)	187.0	11.3
^{252}Cf	2.7×10^{9}	≥750 (RC)	185.7	11.6
^{254}Cf	5.2×10^{6}	≥145 (RC)	186.9	11.8
^{256}Cf	7.4×10^{2}	Asymm. (SS)	189.8	14.6
^{253}Es	2.0×10^{13}	326 (RC)	191	13.4
^{246}Fm	1.2	Asymm. (SS)	199	14.8
^{248}Fm	38	Asymm. (SS)	198	14.5
^{254}Fm	2.0×10^{7}	60 (RC)	195.1	11.7
^{256}Fm	1.0×10^{4}	12 (SS)	197.9	14.4
^{257}Fm	4.1×10^{9}	≈1.5 (SS)	197.6	15.3
^{258}Fm	3.8×10^{-4}	Symm. (SS) FWHM=8	238±3*	14
^{259}Fm	1.5±0.3	Symm. (SS) FWHM=11	242±6*	21
^{259}Md	5.7×10^{3}	Symm. (SS) FWHM=27[c]	≈ 200*	25.5[c]
^{252}No	8.6	Asymm. (SS)	202.4	15.4
1.5-s from ^{248}Cm + ^{18}O	1.5±0.2	Symm. (SS) FWHM=12	234±2*	20.5

[a] Peak-to-valley ratios from radiochemical (RC) or solid-state (SS) measurements.

[b] These are average values of the pre-neutron emission TKE's except for those designated by * which are most probable values.

[c] Private communication, John Wild, 1980.

FIGURES

1. Pre-neutron emission mass-yield curves for ^{254}Fm, ^{256}Fm, ^{257}Fm, ^{258}Fm, and ^{259}Fm. The solid curve for ^{256}Fm is a pre-neutron emission curve while the dashed curve is a provisional mass analysis for ^{256}Fm measured in the same experimental set-up as used for ^{258}Fm (figure from Ref. 9).

2. Pre-neutron emission mass-yield curves for ^{246}Fm (383 events) and ^{248}Fm (74 events) (from Ref. 6).

3. Pre-neutron emission mass-yield distributions for ^{250}Cf, ^{252}Cf, ^{254}Cf, and ^{256}Cf. The data for ^{254}Cf and ^{256}Cf were analyzed in 5 AMU mass bins using an empirical neutron correction similar to that for ^{252}Cf (figure from Ref. 9).

4. $\overline{TKE}$ vs. $Z^2/A^{1/3}$ for heavy actinide isotopes. Solid line represents linear fit of Viola; dashed line is from Unik *et al*. The data for ^{258}Fm and ^{259}Fm are most probable TKE's (figure from Ref. 9).

5. Contour plots of TKE vs. mass fraction for ^{256}Fm, ^{257}Fm and ^{259}Fm (figure from Ref. 10).

6. Pre-neutron emission mass-yield distributions for ^{259}Md and ^{259}Fm. (Data from Refs. 7 and 10.)

7. Pre-neutron emission TKE distributions for ^{259}Md and ^{259}Fm. (Data from Refs. 7 and 10.)

8. Experimental values of $\bar{\nu}_T$ as a function of A of the compound nucleus. Data for SF are shown by +. Measurements for $\bar{\nu}_T$ for (n,f) fission have been corrected to zero excitation energy using $d\bar{\nu}_T/dE_x = 0.11\ MeV^{-1}$ and are shown by o (figure from Ref. 9).

9. $P_t(\nu)$ for ^{250}Cf, ^{252}Cf, and ^{257}Fm for the fission events having the highest TKE's (figure from Ref. 9).

10. Isotopic distributions measured for 98-MeV ^{16}O and 97-MeV ^{18}O bombardments of ^{248}Cm. ^{16}O data are open symbols; ^{18}O data are solid symbols (figure from Ref. 12).

11. Isotopic distributions measured for 115-MeV ^{20}Ne and 116-MeV ^{22}Ne bombardments of ^{248}Cm. ^{20}Ne data are open symbols; ^{22}Ne data are solid symbols (figure from Ref. 12).

12. Energy comparison for heavy product from bombardment of ^{248}Cm with ^{48}Ca ions at the coulomb barrier (figure from Ref. 11).

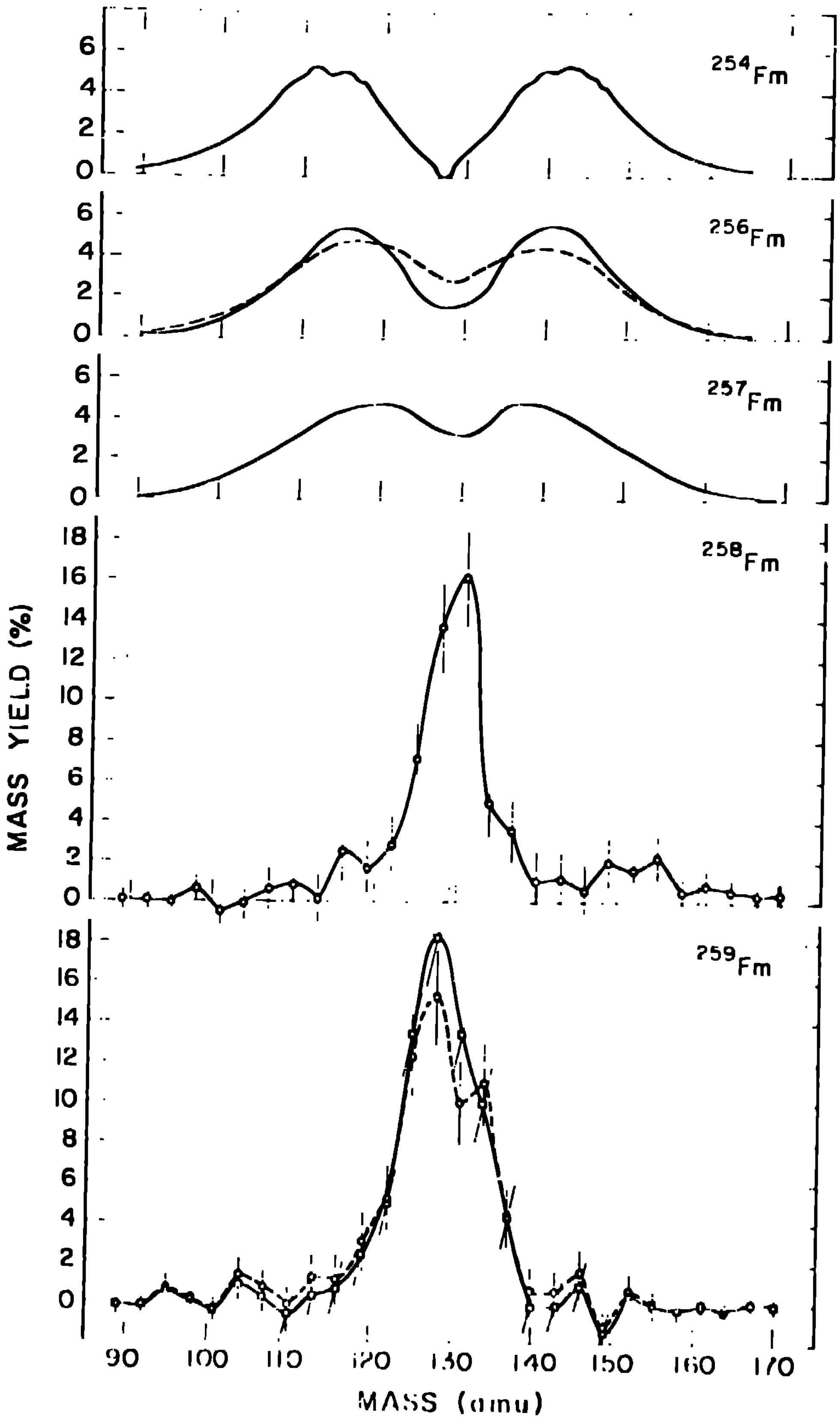
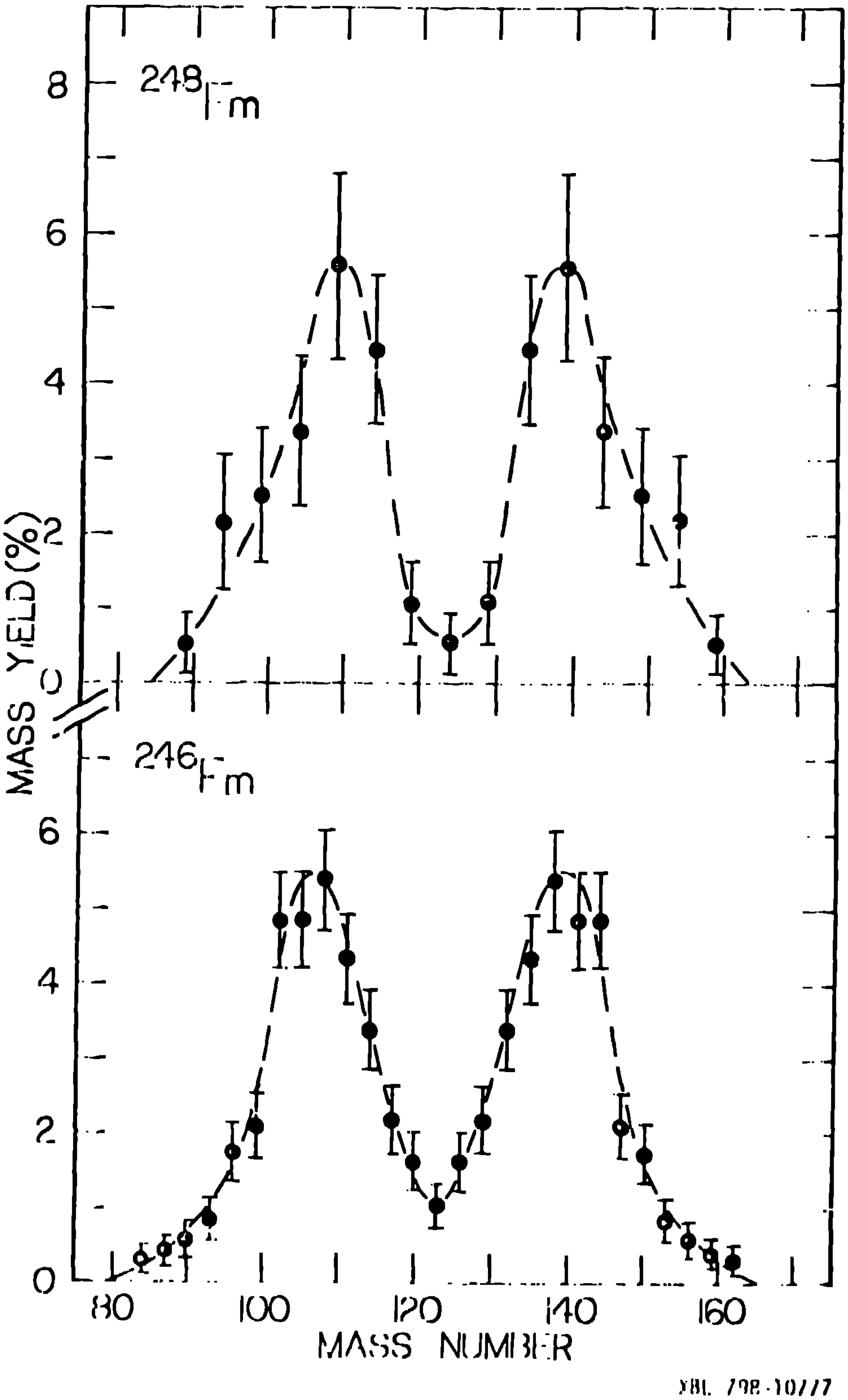


Fig. 1



MASS YIELD (%)
^{248}Fm
^{246}Fm
MASS NUMBER
8
6
4
2
0
6
4
2
0
80
100
120
140
160
XBL 798-10777

Fig. 2

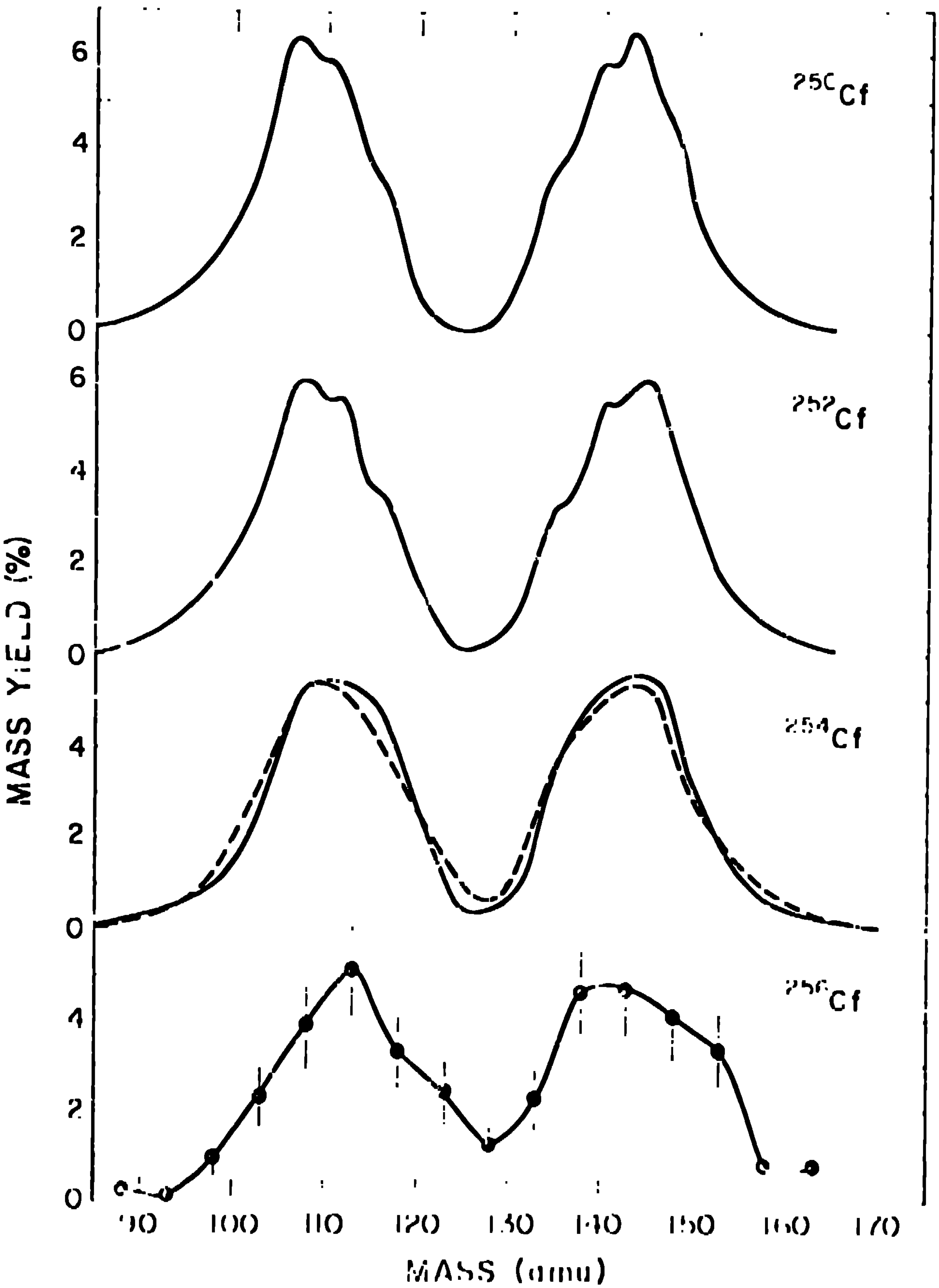
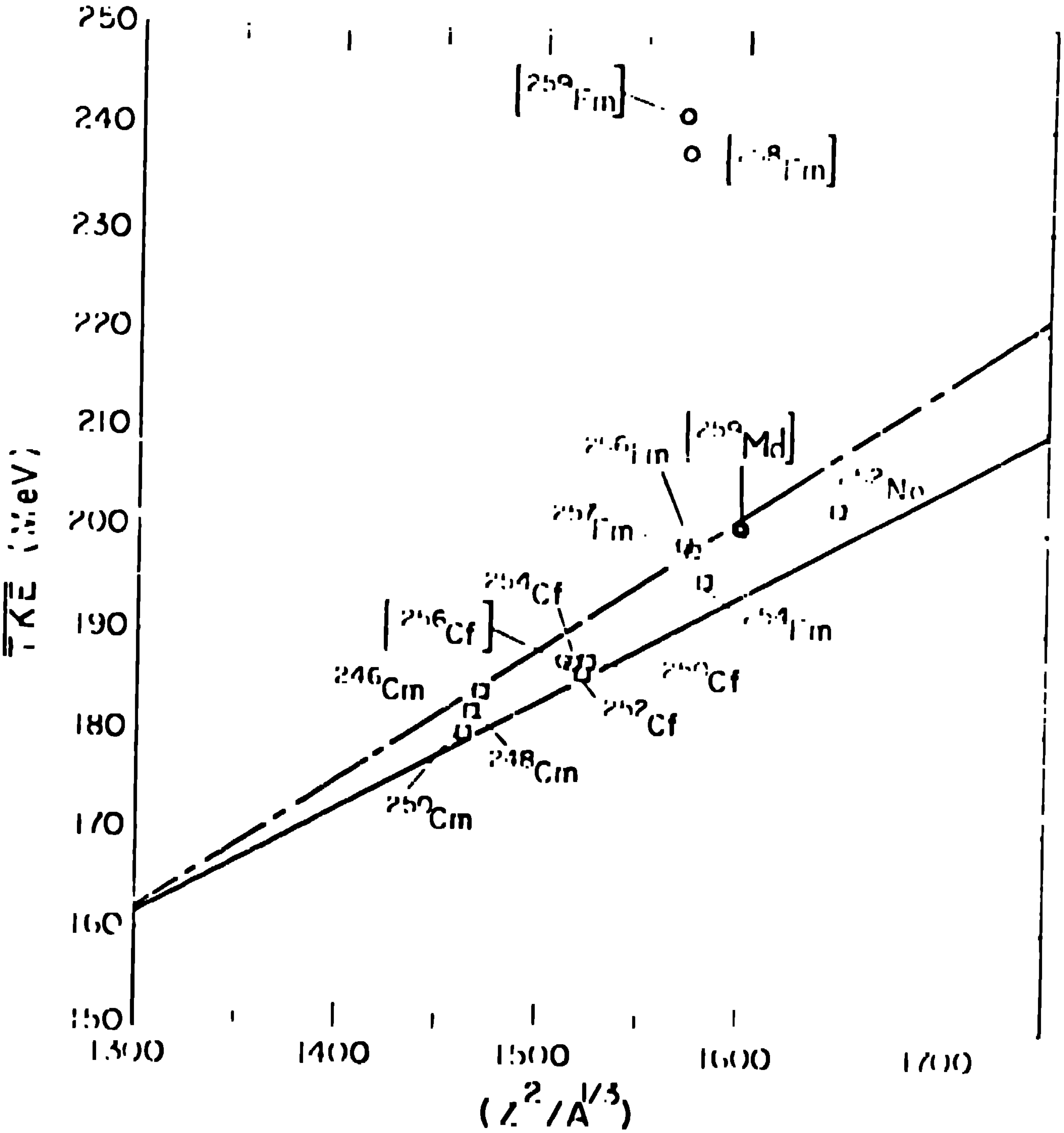

250Cf
252Cf
254Cf
256Cf
MASS YIELD (%)
6
4
2
0
90 100 110 120 130 140 150 160 170
MASS (amu)

Fig. 3



250
240
230
220
210
200
190
180
170
160
150
1300
1400
1500
1600
1700
TKE
(Z²/A^{1/3})
Md
246Cm
248Cm
256Cf
252Cf
Cf
Fm
Fm
Cm
No

Fig. 4

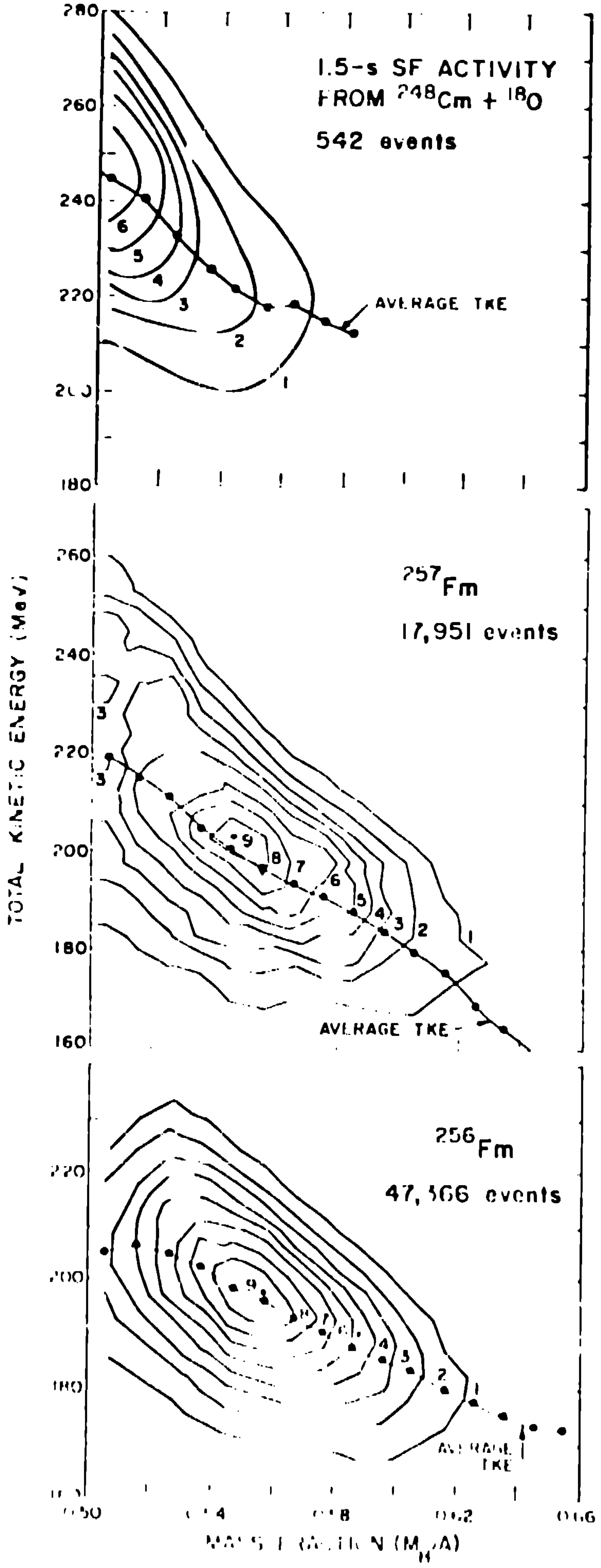

1.5-s SF ACTIVITY FROM $^{248}Cm + ^{18}O$
542 events
AVERAGE TKE
^{257}Fm
17,951 events
AVERAGE TKE
^{256}Fm
AVERAGE TKE
TOTAL KINETIC ENERGY (MeV)

Fig. 5

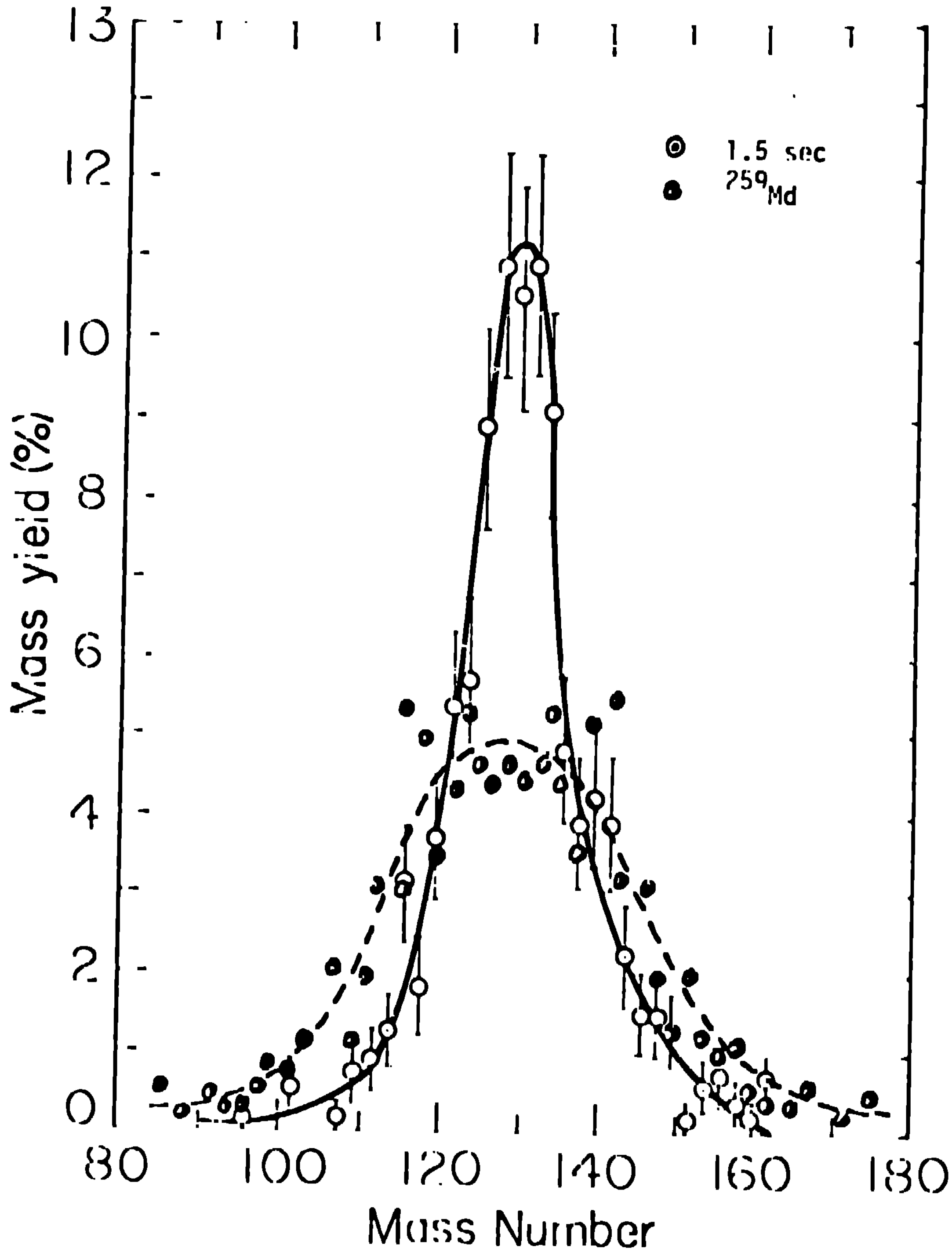
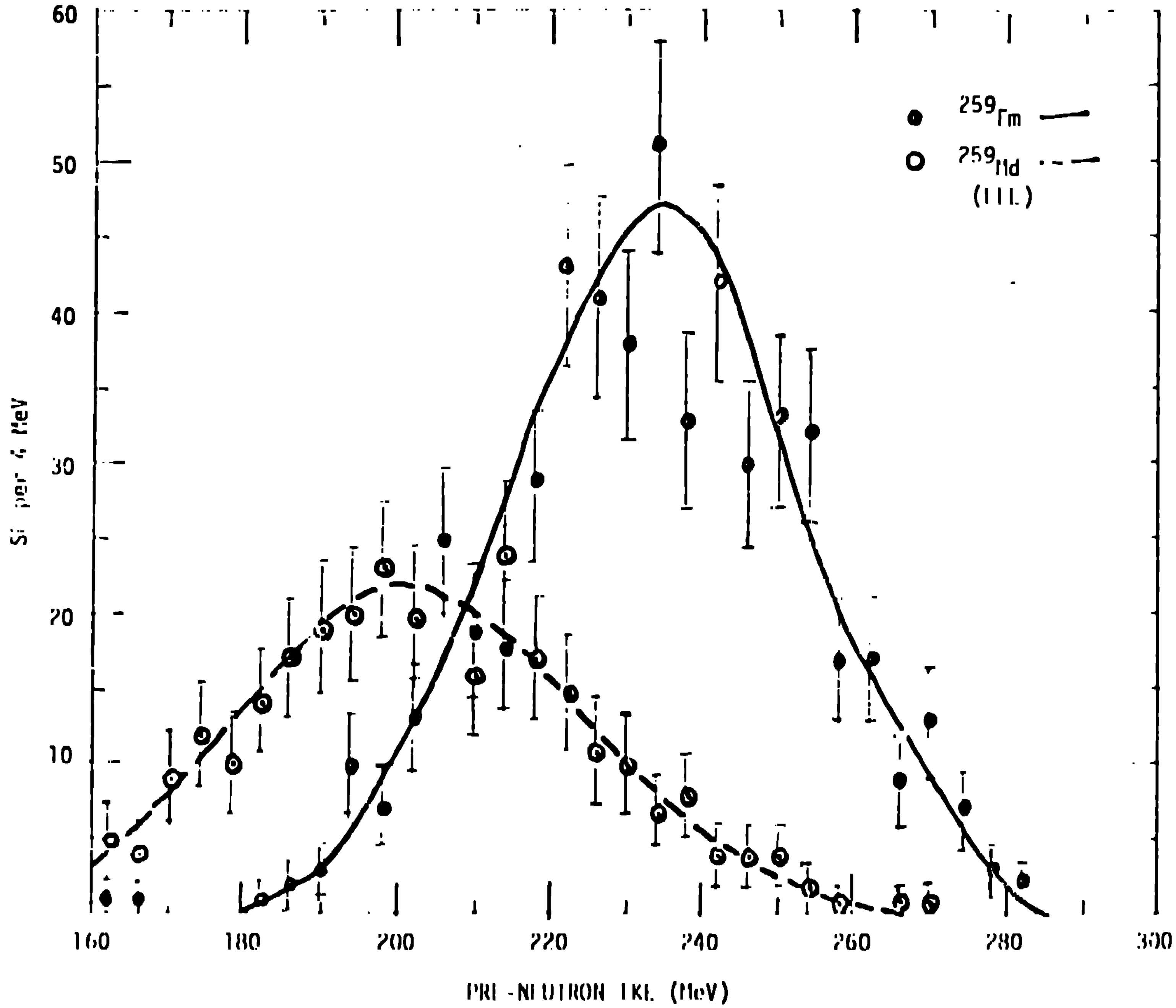
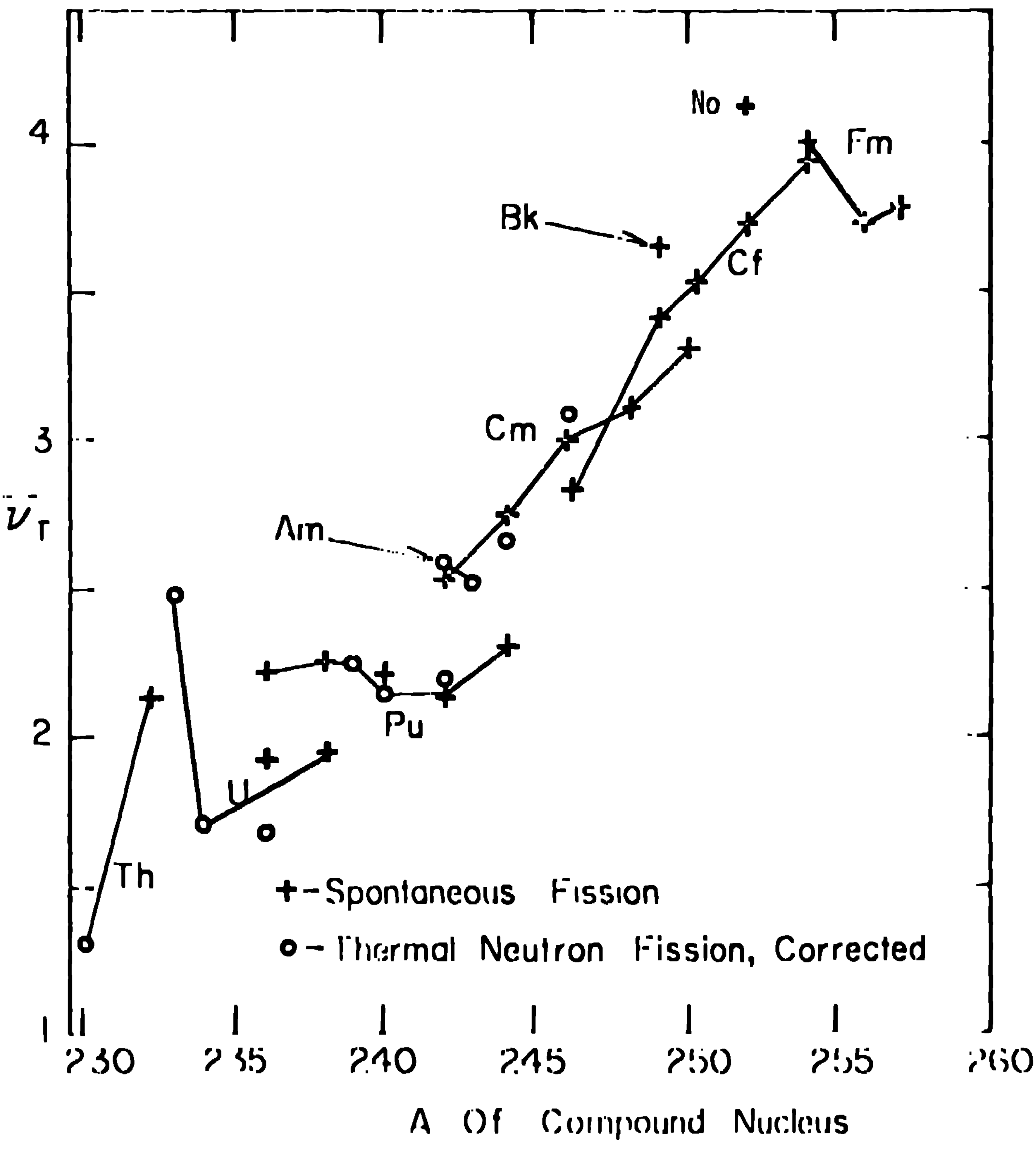

1.5 sec
259Md
Mass yield (%)
Mass Number
0
2
4
6
8
10
12
13
80
100
120
140
160
180

Fig. 6



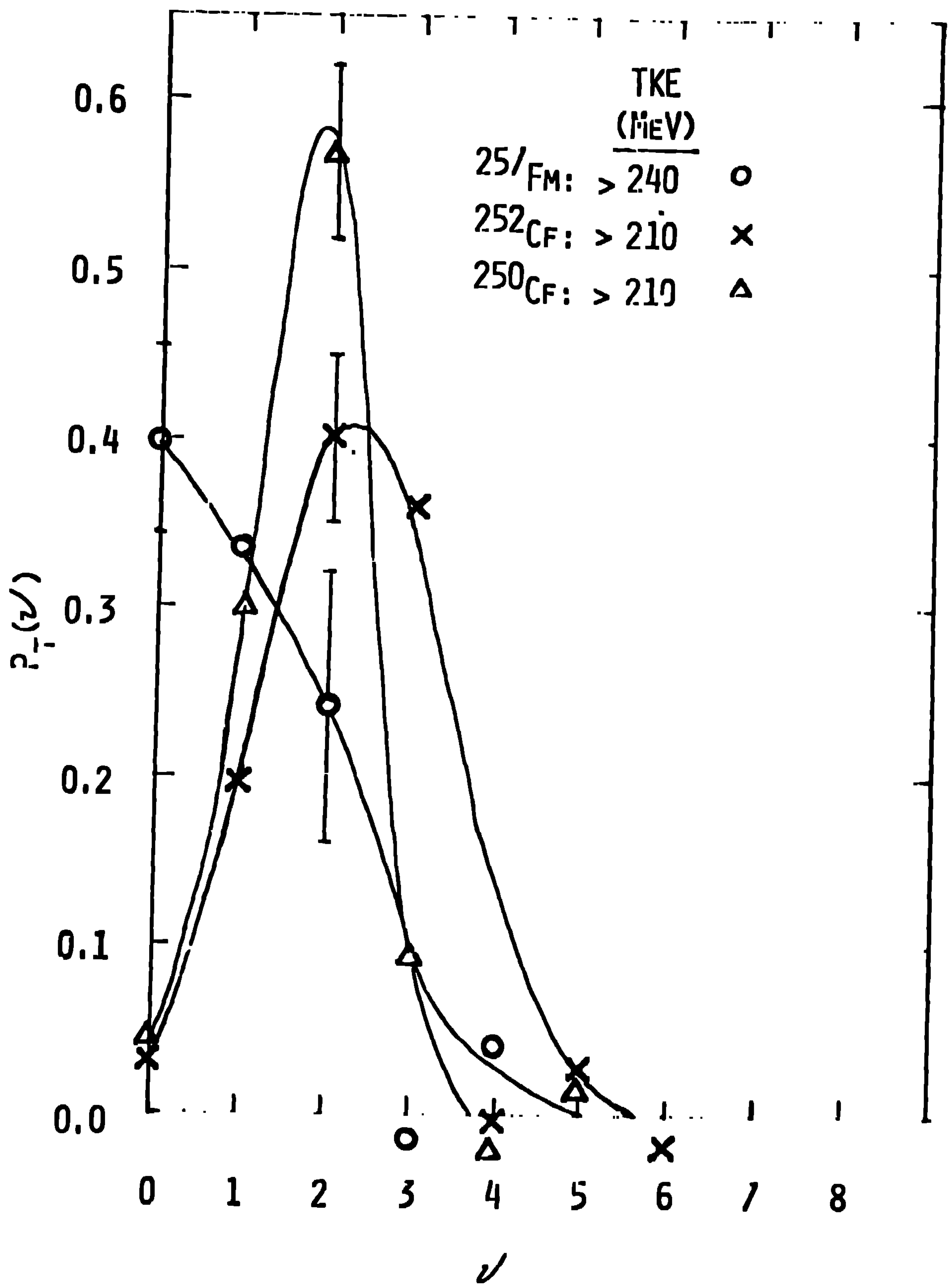
60
50
40
30
20
10
160
180
200
220
240
260
280
300
^{259}Fm
^{259}Md
(ILL)
PRE-NEUTRON TKE (MeV)

Fig. 7



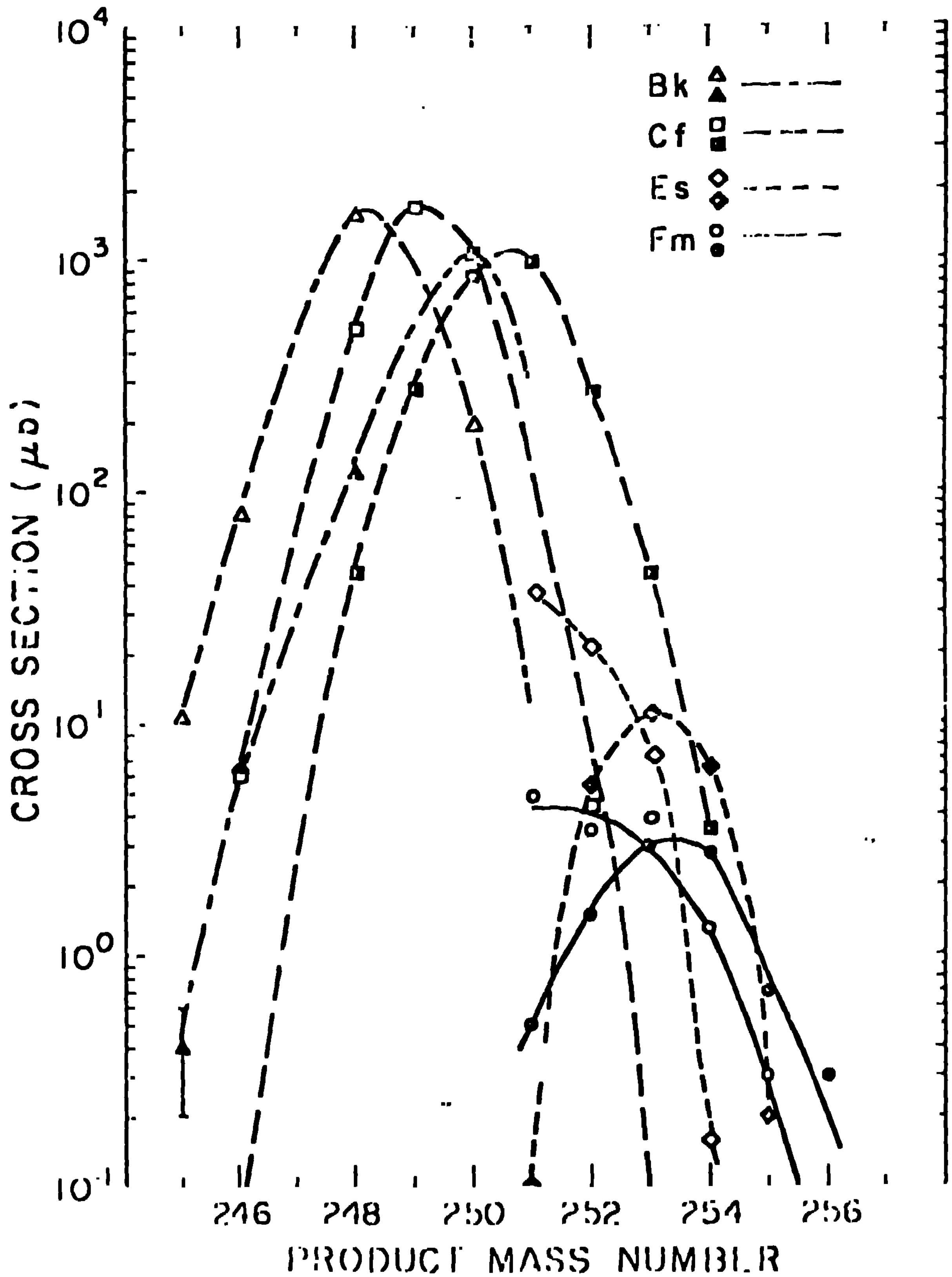
No
Fm
Bk
Cf
Cm
Am
Pu
U
Th
4
3
2
1
$\bar{\nu}_T$
230
235
240
245
250
255
260
A Of Compound Nucleus
+ – Spontaneous Fission
o – Thermal Neutron Fission, Corrected

Fig. 8



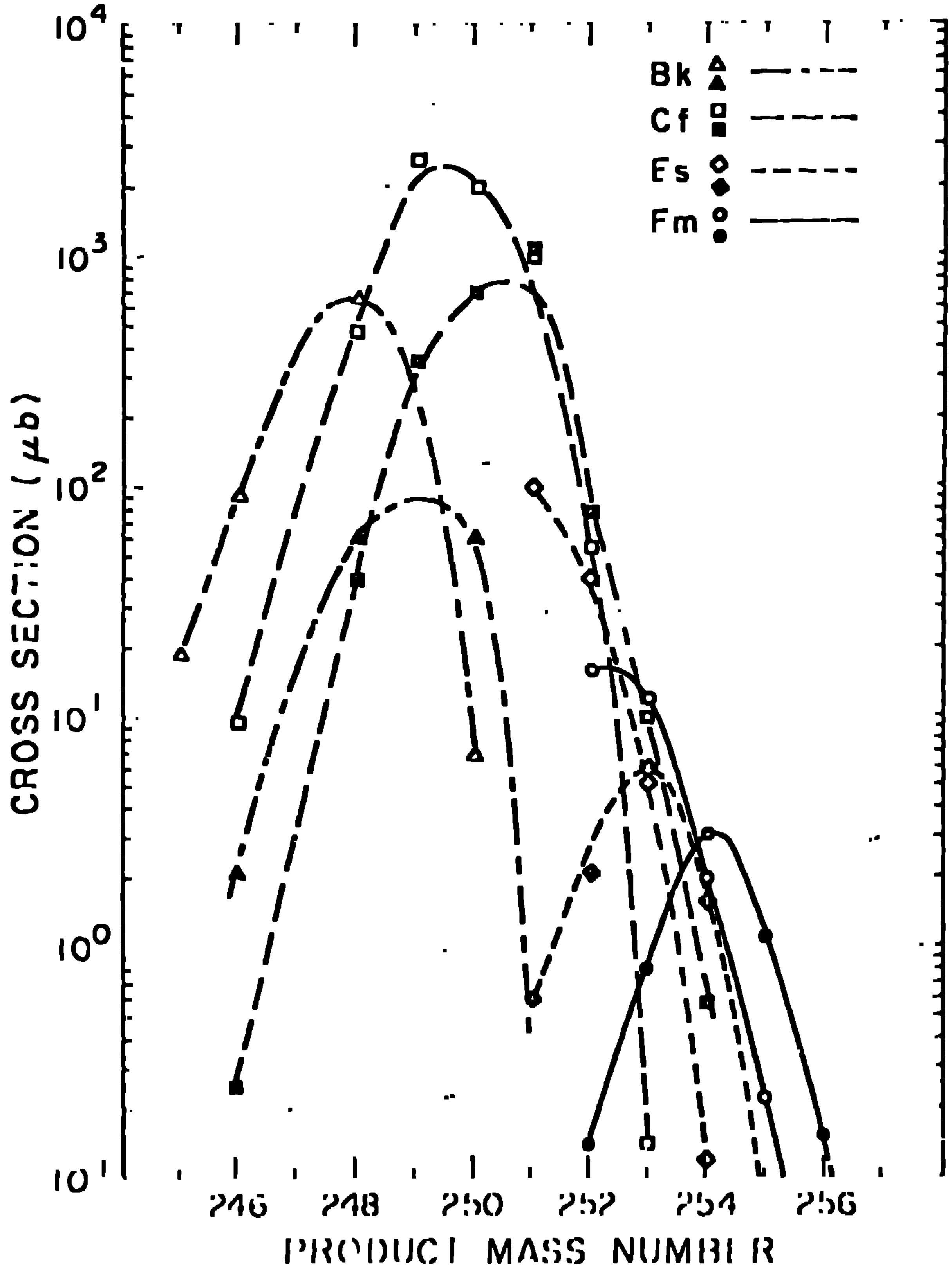
TKE (MeV)
257Fm: > 240
252Cf: > 210
250Cf: > 210
0.6
0.5
0.4
0.3
0.2
0.1
0.0
0 1 2 3 4 5 6 7 8

Fig. 9



Bk
Cf
Es
Fm
CROSS SECTION (μb)
10⁴
10³
10²
10¹
10⁰
10⁻¹
246
248
250
252
254
256
PRODUCT MASS NUMBER

Fig. 10



Bk
Cf
Es
Fm
CROSS SECTION (μb)
10⁴
10³
10²
10¹
10⁰
10⁻¹
246
248
250
252
254
256
PRODUCT MASS NUMBER

Fig. 11

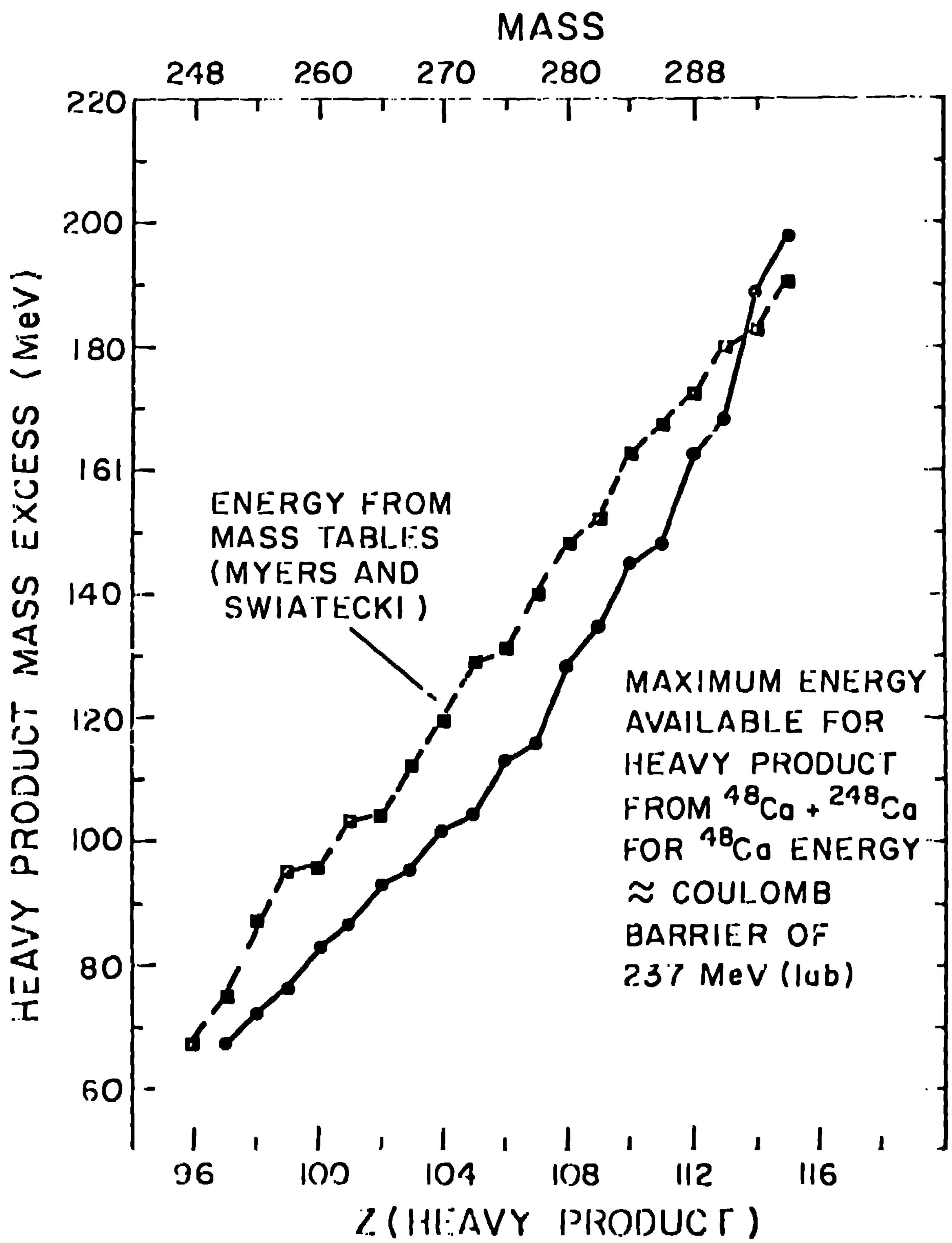

MASS
248
260
270
280
288
220
200
180
161
140
120
100
80
60
HEAVY PRODUCT MASS EXCESS (MeV)
ENERGY FROM
MASS TABLES
(MYERS AND
SWIATECKI)
MAXIMUM ENERGY
AVAILABLE FOR
HEAVY PRODUCT
FROM 48Ca + 248Ca
FOR 48Ca ENERGY
≈ COULOMB
BARRIER OF
237 MeV (lab)
96
100
104
108
112
116
Z (HEAVY PRODUCT)

Fig. 12